

VALENCE BAND SATELLITE IN THE PHOTOEMISSION SPECTRA OF THIN DYSPROSIUM FILMS

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Photoemission spectra of the valence band of dysprosium, taken in the ultraviolet energy range, exhibit a characteristic satellite structure at 5.9 eV below the Fermi edge. This feature is a result of multielectron excitations. This structure can not be identified as a two hole final state in the 4f shell. The experimental observation strongly supports an interpretation of this feature in terms of a many electron shake up process to the unoccupied 4f level.

1. Introduction

Many observations of satellites were reported for photoemission spectra of 3d-metals¹⁻⁴⁾ and the early rare earths^{5,6)}. In the case of nickel the origin of a satellite has been described with a small number of 3d holes or with the completely filled 3d band⁷⁻¹⁰⁾.

The purpose of this paper is to present the result of my recent measurements of angle resolved photoemission from thin (1—10 monolayer) dysprosium films on Si(111), Cu(100) and on fcc Iron thin films deposited on Cu(100). Particular emphasis is placed upon the investigation of a valence band satellite in Dy. The experimental results demonstrate that the resonant 5.9 eV satellite in the photoemission spectra is from dysprosium. This photoemission feature is similar in many respect to the multielectron satellite observed with nickel, but is a consequence of a different multielectron mechanism.

2. Experimental procedure

The experiments were performed in an ultra-vacuum (UHV) system equipped with a hemispherical analyzer with an angular acceptance window $\pm 1.5^\circ$ for angle resolved photoemission as well as a retarding field analyzer for low energy electron diffraction (LEED). The vacuum system was pumped by a combination of turbomolecular, ion and titanium sublimation pumps, and was equipped with a residual gas analyzer and usual pressure gauges.

The light source for these experiments was the 800 MeV ring at the University of Wisconsin-Madison Synchrotron Radiation Center (SRC). The photoemission experiments were carried out on the 3 and 6 meter toroidal grating and the Mark V Grasshopper monochromator. We have acquired photoemission spectra for photon energy between 20–160 eV.

The Cu(100) surface was cleaned by 2 keV Argon ion sputtering and subsequent annealing; a (1×1) LEED pattern was observed. The mounting and cleaning procedure of Si(111) is described in details elsewhere¹¹⁾. A well outgassed Dy source was used to deposit Dy metal on the Si(111), Cu(100) and Fe substrates. During the evaporation, the base pressure in the system was raised to a pressure between 1.3×10^{-8} to 8×10^{-8} Pa. The thickness of the films was determined using a quartz crystal thin film monitor. The deposition rate was typically 10^{-10} meter min^{-1} . The incidence angle θ of the light was defined with respect to the surface normal. s polarized light was defined with the electric vector potential almost completely parallel to the surface ($\theta = 30^\circ$). p polarized light had a large component of the vector potential normal to the surface ($\theta = 64^\circ$). Photoelectrons were collected normal to the surface. Fig. 1 defines the angular parameters used in describing polarization, and light incidence angles.

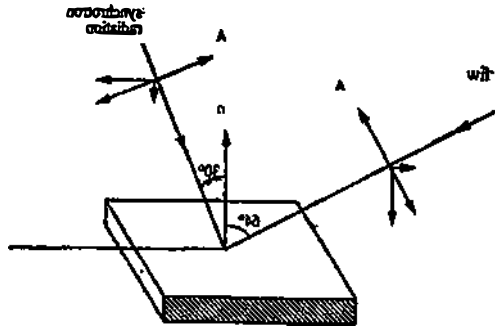


Fig. 1. The angular parameters used in describing the light polarization. When light incidence angle is 30° the greater component of the vector potential A is oriented parallel to the surface (s-polarization). In case of 64° incidence angle, vector potential of the radiation is oriented perpendicular to the surface (p-polarization). n defines the sample normal vector.

3. Results and discussion

Fig. 2 shows the photoemission spectra of 2.5 monolayers (ML) of Dy deposited on Fe and 6 ML Dy on Cu(100). The spectra were taken at 50 eV photon

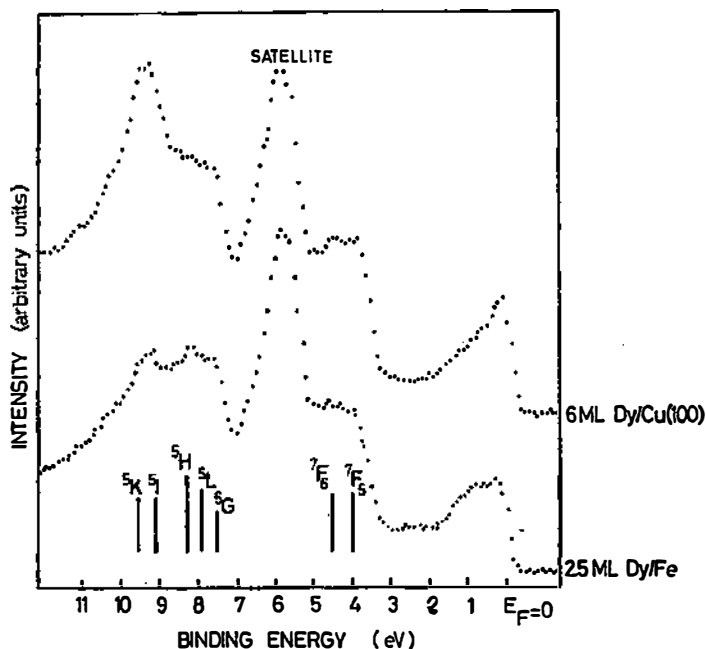


Fig. 2. Photoemission spectra of dysprosium overlayers on Cu (100) and Fe. All spectra are taken with 50 eV photon energy and normal emission. Binding energies referenced to the Fermi energy. The dysprosium overlayer thickness, in monolayers (ML) is noted.

energy with p-polarized light. There is a wealth of dysprosium valence band features, in part a result of the different 4f multiplets¹²⁻¹⁴). The various 4f multiplets are the $7F_5$ and $7F_6$ levels (at 4.1 ± 0.1 eV), $5L$ and $5G$ levels (at 7.9 ± 0.1 eV), $5I$ and $5H$ levels (at 9.0 ± 0.2 eV) and the $5K$ level (at 10.0 ± 0.2 eV). The photoemission intensity near the Fermi energy for dysprosium on Cu(100) is a result of the dysprosium 5d bands. For dysprosium on Fe, the peaks near the Fermi edge are due to combined iron 3d and dysprosium 5d-6s bands. The emission at (5.9 ± 0.2) eV below Fermi energy was not expected on the basis of the initial dysprosium density of states¹²⁻¹⁴). This feature has 1.2 eV full width at half maximum (FWHM) for Dy on Cu(100) and 1 eV for Dy on iron. The binding energy of this peak is independent from incident photon energy. Fig. 3 illustrates electron energy distribution curves of 1.3 ML and 6.5 ML Dy on Si(111), spectra taken with s and p polarized light for the 6.5 ML film as indicated in the figure. Dysprosium was deposited onto the room temperature substrate. The 1.3 ML film exhibits a Dy-silicide feature with Dy 5d-Si 3p at (2 ± 0.2) eV and Dy 4f-Si 3sp hybridization at (5.3 ± 0.1) eV energy below Fermi edge. The 6.5 ML Dy films exhibit electronic features similar to those of pure dysprosium. The photoemission intensity of the 5.9 eV feature for the 6.5 ML Dy films is much more intense at an incidence angles of 64° off normal (p-polarized light) than with light incident at an angle of 30° off normal (s-polarized light), as seen in Fig. 3.

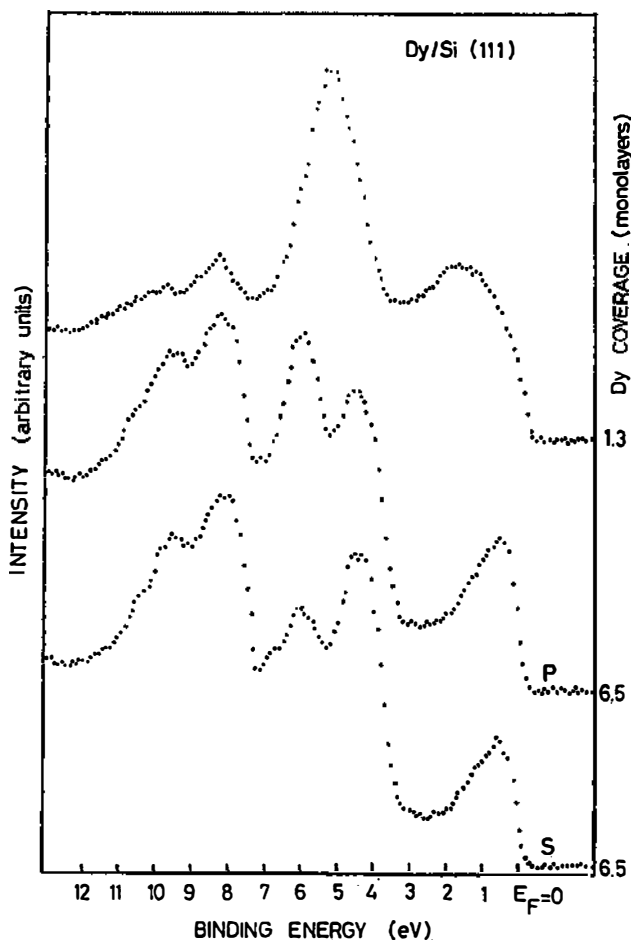


Fig. 3. Electron energy distribution curves (EDC's) of dysprosium valence band on Si(111). The incident light polarization s ($\theta = 30^\circ$), and p ($\theta = 64^\circ$) are shown in the figure for 6.5 ML Dy coverage. Spectra were taken with 50 eV photon energy and normal emission.

In order to determine the origin of this 5.9 eV feature we have to consider several possible mechanisms as cause for the observed satellite. The oxygen (2p) feature might be expected to be observed at 5.9 eV binding energy as a result of oxygen contamination. The study of oxygen exposure to gadolinium gave a reported O (2p) peak at 5.8 to 6.0 eV below E_F and 3 eV FWHM¹⁵⁾. This FWHM value is much greater than observed with our freshly deposited dysprosium. Furthermore the photoemission intensity of the 5.9 eV feature increases with p-polarized light with respect to the 4f bands. It is very unlikely that oxygen would induce no O (2p) features of p_x or p_y character. It can be seen in Fig. 3 that the Dy 5d intensity increases with increasing light incidence angle. Therefore we would expect the 5.9 eV photoemission feature and the 5d band share the same $3_{2^2-1/2}$ symmetry character. The origin of this peak related with the Dy 5d band.

The satellite photoemission feature observed for the dysprosium thin films has fixed binding energy. Thus, this peak is not a result of a characteristic Auger decay process. No significant features with constant electron kinetic energies are observed with photon energies between 30 to 60 eV. The other possibility for this peak can be a surface state of dysprosium, but all deposited films were disordered. The 5.9 eV feature is present for all films on Cu, Si and Fe, suggesting no interaction between dysprosium and the substrate.

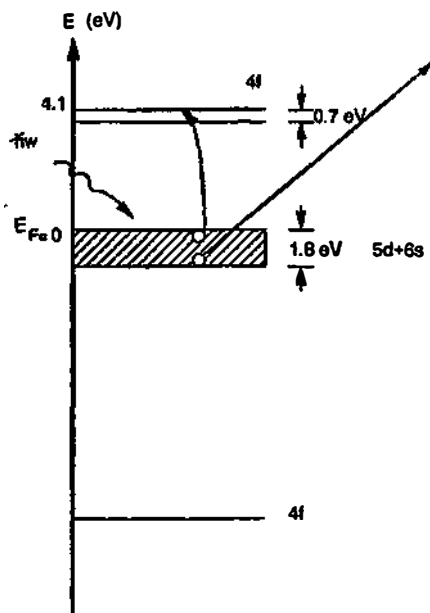


Fig. 4. Schematic representation of the multielectron shake-up process. The outgoing electron from the bottom of the conduction band can shake-up an electron at the top of the conduction band to the unoccupied 4f levels.

The unoccupied 4f levels of dysprosium have been determined at 4.1 eV above the Fermi energy by Lang et al. using bremsstrahlung isochromat spectroscopy (BIS)¹²⁾. An electron excitation from the 5d band to the unoccupied 4f levels accompanying photoemission from the 5d6s band would result in a satellite feature. This multielectron shake-up photoemission process is shown schematically in Fig. 4. The binding energy of this satellite would be the energy of the 5d to 4f interband transition, plus the binding energy of the photoemitted electron. Note that the photoemitted electron comes from bottom of the Dy 5d band (Fig. 4). The binding energy of the photoemitted electron from the initial state will be about 1.8 eV. As a result of the interband transition the satellite will have 5.9 eV binding energy (4.1 plus 1.8 eV) consistent with the observed 5.9 eV peak. This interpretation is supported by the variation of the satellite intensity with light polarization as seen in Fig. 3. The photoemission intensity of the dysprosium 5d band and the 5.9 eV feature increased with p-polarized light. Both are sharing the same final state character in photoemission.

In Fig. 3 we have another evidence that a d to f shake-up process is occurring and it requires an initial state 5d occupancy. The result obtained for the 1.3 ML Dy films on Si(111) shows Dy silicide structure and apparently has different chemical properties. This film doesn't show a satellite feature. It is clear that inter-diffusion of Dy atoms and silicide formation reduced the coordination number of Dy. This effectively changes the density of states near the Fermi level. In case of 1.3 ML coverage, dysprosium has an atomic character with very low 5d electron occupation¹⁶⁾, which effectively closes the shake-up excitation channel. When the Dy thickness is increased to 6.5 ML a 5d occupancy has fully developed and an accompanying satellite feature at (5.9 ± 0.2) eV appears.

In summary, a valence band satellite photoemission feature of dysprosium thin films has been observed and has been assigned to a multielectron shake-up process resulting from a transition from the 5d band to the unoccupied 4f level. The shake-up resonance is present only in the ultraviolet energy range (30—60 eV) with enough 5d occupation. The feature is very weak below 30 eV and is not observed above 60 eV photon energy.

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SATELITI VALENTNE VRPCE U SPEKTRU FOTOEMISIJE TANKIH
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Spektri UV fotoemisije valentne vrpce Dy pokazuju karakterističnu strukturu 5,9 eV ispod Fermi nivoa. Ta pojava je rezultat višeelektronskih pobuđenja. Tu strukturu nemoguće je objasniti konačnim stanjem s dvije šupljine u 4f ljusci. Eksperimentalni rezultati daju jaku podršku objašnjenju pomoću mnogoelektronskih shake-up procesa u nepopunjeni 4f nivo.